Magnetic properties of an Fe/Cu granular multilayer

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A Cu/Fe granular film, formed from a multilayer film and composed of particles of Fe imbedded in Cu, has had several of its important properties investigated. Measurements include ferromagentic resonance, magnetoresistance, Mössbauer effect, magnetic viscosity, and magnetization. The two-phase behavior of these immiscible alloy systems, and the effect of multilayering on the shape of the magnetic precipitates, can explain some of these properties. An explanation of the ferromagnetic resonance line shape is proffered. An extraordinary macroscopic quantum tunneling effect is found to govern the magnetic relaxation at the lowest temperatures.

I. MAGNETORESISTANCE

We have manufactured a Cu/Fe multilayer, [Cu(50 Å)/Fe(10 Å)]×50 (and a Cu/Dy multilayer [Cu(100 Å)/Dy(40 Å)]) on Kapton. The samples were prepared by thermal evaporation in a high-vacuum system. Due, in part, to the low miscibility of Fe in Cu, this process can produce samples with small granular particles of Fe imbedded in the Cu matrix, instead of the usual layered structures. A similar process was recently used by IBM to produce their discontinuous (granular) $Ni_{80}Fe_{20}$/Ag multilayer films which currently hold the record for the largest sensitivity in any giant magnetoresistive (GMR) structure.[1] Although our Cu/Fe has only modest GMR, and Dy/Cu has almost none, our study of granular multilayers should provide insights into the magnetic structure of $Ni_{80}Fe_{20}$/Ag.

The 4.2 K magnetoresistance of the Fe/Cu granular multilayer is plotted as a function of external applied magnetic field in Fig. 1, along with the 4.2 K magnetic moment of the sample, obtained with a superconducting quantum interference device magnetometer. The magnetoresistance is large, decreases monotonically with field, and is independent of the field direction. These are the telltale characteristics of GMR, a phenomenon often found in multilayers and granular materials. GMR is associated with magnetically inhomogeneous material, and is caused by the magnetic scattering of conduction electrons by the nonaligned magnetic entities.[2] At 4.2 K and 60 kOe, the fractional change of resistance $\Delta\rho/\rho = -15\%$, but neither the magnetoresistance nor the magnetic moment is completely saturated yet. This failure to saturate is not due to ordinary magnetic hardness, but is caused by the existence of a paramagnetic component, most likely Fe singlets, dimers, or trimers dissolved within the Cu matrix, which coexist with the Fe precipitates. Further evidence for the existence of a paramagnetic iron moment in the as-deposited Fe/Cu film will become apparent in the Mössbauer effect results, presented in Sec. IV.

Unlike Fe/Cu, the Dy/Cu sample showed only ordinary magnetoresistance, characterized by both a nonmonotonic field dependence and a sensitivity to the magnetic-field direction.

II. FERROMAGNETIC RESONANCE

We have obtained the ferromagnetic resonance (FMR) spectrum of the Cu/Fe granular multilayer sample at 9.55 GHz with the magnetic field both parallel and perpendicular to the film plane. (The Dy/Cu spectrum was not detected.) The spectra depend hardly at all on temperature, and the room-temperature parallel and perpendicular absorption-derivative spectra are shown in Fig. 2. The parallel resonance is centered at 1.43 kOe and has a linewidth $\Delta H_{\parallel}=0.975$ kOe, while the perpendicular resonance is centered at 7.87 kOe with a linewidth $\Delta H_{\perp}=1.95$ kOe. Kittel[3] has derived the fields for resonance in the parallel and perpendicular configuration of thin magnetic films, H. In the absence of magnetic anisotropy they are given by the equations

$$f/\gamma=(H)^{1/2}(H+4\pi M)^{1/2} \quad \text{(parallel resonance)},$$

$$f/\gamma=H-4\pi M \quad \text{(perpendicular resonance)}, \tag{1}$$

where f is the microwave frequency, γ is the gyromagnetic ratio, and M is the magnetization. (For a pure iron film, γ=3.2 GHz/kOe and $4\pi M$=21.5 kOe.)

Using the Kittel equations for the Cu/Fe sample, we obtain g=2.18 and $4\pi M$=4.87 kOe using the data shown in

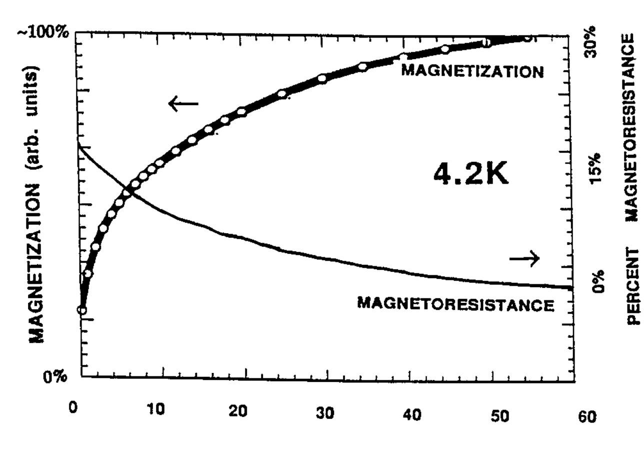

FIG. 1. Magnetization and magnetoresistance vs magnetic field at 4.2 K for Cu(50 Å)/Fe(10 Å). An easily saturable ferromagnetic component and a hard to saturate paramagnetic component are evident.

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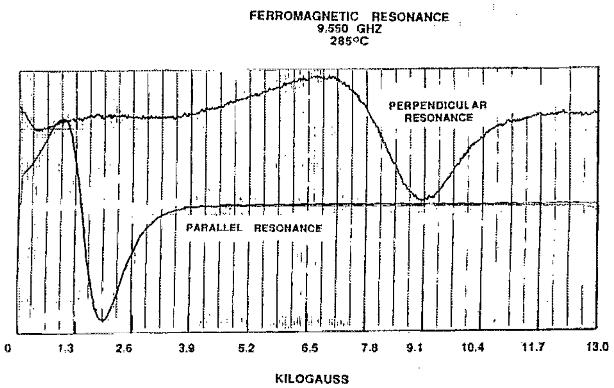

FIG. 2. 9.55 GHz FMR spectrum of Cu(50 Å)/Fe(10 Å) with applied field parallel and perpendicular to plane of film.

Fig. 2. The low value of magnetization results from the low value of the iron filling factor, ≈20%. Griscom *et al.*[4] have shown that it is proper to use the average value of the magnetization in the Kittel equations for some granular or precipitated films. Our derived values are in agreement with this conclusion, and this result is further justified below.

In a previous publication[5] one of the authors has shown that thin films of granular $Fe_{20}Cu_{80}$, made by coevaporation of the elements, with no attempt at multilayering, have larger FMR linewidths than those shown in Fig. 2. As-deposited films have linewidths of 2.5 kOe, and films annealed above 500 °C have linewidths of 4.0 kOe. The as-evaporated linewidth is attributed to a completely random distribution in the direction of the anisotropy fields[6] of the individual single-domain iron precipitates, resulting in a powder-pattern FMR lineshape with a linewidth $\Delta H=(5/3)(2K/M)$. Iron precipitates in thin films annealed at temperature higher than 500 °C have grown into multidomain particles with a linewidth[4] $\Delta H=(0.7)(4\pi M/3)$. The smaller linewidths of the granular multilayer—especially that of the parallel resonance—shows that the layer formation has restricted the spread in particle orientations, constraining it to be a less-than-random distribution. In the following section, we present some ideas on this matter, which also serve to explain why $\Delta H_\perp$ is so much larger than $\Delta H_\parallel$.

III. NETZELMANN'S CONJECTURE

The underlying multilayer structure can distort the shape of the iron precipitates by (a) inducing uniaxial distortions in what, otherwise, would be a sphere, or (b) produce a collection of pancakelike magnetic particles partially aligned along the layers. These alternatives are the two extremes, neither of which may prevail in practice. The high-sensitivity NiFe-Ag granular multilayers produced by the IBM group[1] belong to group (b).

We now assume that the sample is a collection of nearly flat islandlike particles—a discontinuous multilayer. (Assuming that the particles are spheroids aligned parallel to the layer direction does not alter our quantitative conclusions, and can be treated in the same manner.) Each particle is characterized with a demagnetization tensor N_p, and the thin

TABLE I. Values for Kittel demagnetizing factors.

Effective demagnetizing factor	Prependicular resonance	Parallel resonance
N_z	$4\pi f+4\pi(1-f)(1-2\epsilon)$	$4\pi 1-f)\epsilon$
N_x	$4\pi(1-f)\epsilon$	$4\pi f+4\pi(1-f)(1-2\epsilon)$
N_y	$4\pi(1-f)\epsilon$	$4\pi(1-f)\epsilon$

film is characterized with a tensor N_t. Netzelmann[7] has proposed that the effective magnetostatic energy density F_N be approximated as

$$F_N=\tfrac{1}{2}(1-f)\mathbf{M}\cdot\mathsf{N}_p\cdot\mathbf{M}+\tfrac{1}{2}(f)\mathbf{M}\cdot\mathsf{N}_t\cdot\mathbf{M}, \tag{2}$$

where f is the volumetric filling factor of magnetic particles and $\mathbf{M}$ is the particle magnetization. Except for the constraint imposed by Eq. (2), each particle is assumed to precess independently during FMR. Netzelmann[7] and Yu, Harrell, and Doyle[8] have used this approach to determine particle orientation distributions from FMR spectra in magnetic tapes.

The generalized Kittel equation,[3] applicable to an ellipsoid with demagnetizing factors designated by N_x, N_y, and N_z, is

$$f/\gamma=\{[H_0+(N_y-N_z)M_0][H_0+(N_x-N_z)M_0]\}^{1/2}, \tag{3}$$

where the z axis is always in the direction of the applied field H_0, and the y and x axes are perpendicular to z. Choosing diagonal values $\mathsf{N}_p=4\pi(1-2\epsilon,\epsilon,\epsilon)$ and $\mathsf{N}_t=4\pi(1,0,0)$, appropriate to an ellipsoidal particle and a thin film, respectively, we obtain the following values for the Kittel demagnetizing factors, for parallel and perpendicular resonance, as shown in Table 1.

We now assume that the FMR linewidth is primarily caused by a flat distribution of particle ellipticity parameters ϵ, which vary between $\epsilon=0$ and $\epsilon=\epsilon_0$. Using Eq. (3), the perpendicular resonance linewidth is given by $\Delta H_\perp=4\pi M_0\epsilon_0(1-f)$, from which we obtain the value $\epsilon_0=0.12$ from the experimental value of $\Delta H_\perp$. The Netzelmann–Kittel equation yields $\Delta H_\parallel=0.65$ kOe for the value of the parallel resonance linewidth, compared to the experimental value of 0.97 kOe, and shows why the parallel resonance linewidth differs than from the perpendicular resonance linewidth. The equations also show that the appropriate magnetization to use in the Kittel formula is the average global magnetization for both flat and spherical precipitated particles.

Using nearly spherical particles with a distribution of ellipsoidal distortions, instead of pancake-shaped particles, yields the same quantitative conclusions. However, using nearly spherical particles results a larger discrepancy in the ratio of the two linewidths than using nearly flat particles. However, from FMR linewidths alone, we are hesitant to draw too many conclusions. Other[1] granular multilayer systems are found to have flat, islandlike precipitates.

IV. OTHER EXPERIMENTAL RESULTS

Mössbauer effect measurements were obtained at 80 K on the Cu(50 Å)/Fe(10 Å) multilayer studied here. The spectrum consisted of a hyperfine field sextet with splittings appropriate to ferromagnetic a-Fe and a broad zero-field "singlet" which we associate with the paramagnetic irons discussed in connection with Fig. 1. The paramagnetic component is strong, in agreement with magnetoresistance and magnetization results. The Mössbauer measurements indicate that the moments are in plane, as expected for either spheres or platelets with low magnetic anisotropy imbedded in a thin film. Details of another, more magnetically dilute, multilayer have been published elsewhere.[9]

The dynamic properties of this Fe/Cu granular multilayer has been investigated between 1.6 and 16 K by examining the time dependence of the thermoremanent magnetization, otherwise known as the magnetic aftereffect. Below $T^* \approx 4$ K the magnetic relaxation time becomes independent of temperature, which we interpret as evidence of quantum-mechanical tunneling of the single-domain magnetization of the Fe particles. Aharoni[10] has shown that shape anisotropy, as well as crystalline anisotropy, alters the temperature T^* at which quantum tunneling first appears. The present study, which seeks to quantify the shape anisotropy, may influence the interpretation of the quantum tunneling results. Further discussion of this remarkable effect can be found in Ref. 11.

Magnetic properties of similar granular-multilayer Fe/Cu and Dy/Cu films have been published elsewhere.[12]

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